INL/CON-07-12919
PREPRINT

Post-Irradiation Examination of Irradiated Fuel Outside the Hot Cell

European Working Group, Hot Laboratories and Remote Handling

Dawn E. Janney
Adam B. Robinson
Thomas P. O'Holleran
R. Paul Lind
Marc Babcock
Laurence C. Brower
Julie Jacobs
Pamela K. Hoggan

September 2007

Post-irradiation-examination of irradiated fuel outside the hot cell

Dawn E. Janney[*]*, Adam B. Robinson, Thomas P. O'Holleran, R. Paul Lind, Marc Babcock, Laurence C. Brower, Julie Jacobs, Pamela K. Hoggan*
Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415-6188 USA

Abstract
Because of their high radioactivity, irradiated fuels are commonly examined in a hot cell. However, the Idaho National Laboratory (INL) has recently investigated irradiated U-Mo-Al metallic fuel from the Reduced Enrichment for Research and Test Reactors (RERTR) project using a conventional unshielded scanning electron microscope outside a hot cell. This examination was possible because of a two-step sample-preparation approach in which a small volume of fuel was isolated in a hot cell and shielding was introduced during later stages of sample preparation. The resulting sample contained numerous sample-preparation artifacts but allowed analysis of microstructures from selected areas.

KEYWORDS: scanning electron microscopy (SEM), sample preparation, Reduced Enrichment for Research and Test Reactors (RERTR), irradiated fuel

1. Introduction
The Reduced Enrichment for Research and Test Reactors (RERTR) project is developing low-enrichment uranium (<20% U-235) fuels that can be used instead of the current high-enriched uranium fuels in research reactors. Two kinds of fuel are under development: dispersion fuels, in which the uranium is dispersed in fine particles, and monolithic fuels, in which it is a constituent of a homogeneous alloy [1].

This paper describes preparing two samples of irradiated RERTR-6 dispersion fuels for examination in a conventional (unshielded) scanning electron microscope and illustrates consequences of the sample-preparation technique for the kinds of data that can be collected from the samples.

2. Samples
Two dispersion fuel plates were prepared at the Fuels and Applied Sciences Building (FASB) at the Idaho National Laboratory (INL). Each plate had a central fuel zone approximately 0.06 cm thick, and consisting of fine particle (typically <100 μm diameter) of U-7Mo embedded in an aluminum-alloy matrix. The fuel zone was sandwiched between two layers of Alloy 6061 cladding, each 0.04 cm thick. The total nominal as-fabricated thickness of each plate was 0.14 cm.

The plates were irradiated in the Advanced Test Reactor (ATR) at the INL to an average burnup of 49-50% in the mid-plate region. Irradiation ended in November, 2005. A gamma scan performed in September, 2006, indicated that the isotopes with the highest contribution to the sample radioactivity at that time were Ce-144, Nb-95, Zr-95, Ru/Rh 106, Cs-137, Cs-134, Ru-103, and Ce-141 (listed in order of decreasing contribution to the gamma scan); however, some of the shortest-lived isotopes (particularly Nb-95) would have almost completely decayed to stable or long-lived isotopes by the time samples were prepared for microscopy in late May, 2007. Radiation readings from a punching with a nominal 1 mm diameter through the thickness of one of the plates were 300 mSv/hr combined beta-gamma and 3 mSv/hr gamma (as measured at contact with a Ludlum model R020 ion chamber detector) when the samples were prepared for SEM analysis.

Each irradiated plate was prepared for optical metallography in the hot cell by grinding it with a series of abrasives, ending with a nominal 3 μm abrasive polishing fluid. Samples were not etched before examination. Optical examination of the irradiated plates indicated that each retained its layered structure consisting of a central fuel zone with particles dispersed in a matrix, surrounded by two particle-free layers corresponding to the original cladding (Figure 1a). Higher-magnification views (Figure 1b) show that the microstructure of the fuel zone consists of particles, dispersed in a matrix. Each particle has a dark center surrounded by a lighter-colored ring. Previous analyses of similar samples (e.g., [2-6]) indicate that the rounded particles are what remains of the original U-Mo

[*]Telephone: 208-533-7478, FAX: 208-533-7996, e-mail: dawn.janney@inl.gov

alloy particles, the rings around them are U-Al intermetallic reaction products formed during irradiation, and the matrix is an aluminum alloy.

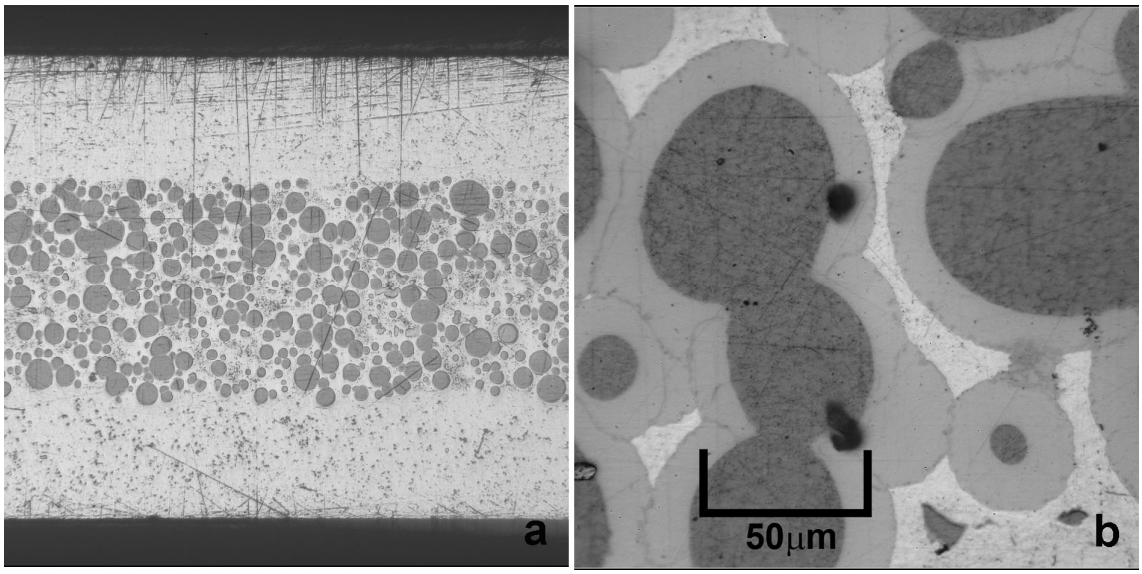

Figure 1: Optical micrographs showing typical microstructures in irradiated fuel. a) Cross-section of entire plate, showing central zone with fuel particles surrounded by particle-free zones. Total thickness of plate is approximately 0.14 cm. b) Detailed view of fuel zone.

3. Sample preparation for electron microscopy

Sample preparation for electron microscopy occurred in two steps. In the first, which was performed in the argon-atmosphere hot cell at the Hot Fuel Examination Facility (HFEF) at the Idaho National Laboratory, a commercially available press that had been modified slightly for easier operation in a hot-cell environment (Figure 2) was used to produce cylindrical punchings with nominal 1 mm diameters. Each cylinder had a nominal height of 1.4 mm (the thickness of the fuel plate). The punchings were placed in individually labeled containers and transferred into the air-atmosphere glove box at the Electron Microscopy Laboratory (EML) at the Idaho National Laboratory.

At the EML, each punching was placed on its side inside a commercially available 1.25 inch (3.175 cm) diameter, 0.75 inch (1.9 cm) tall phenolic ring that had been attached to a piece of duct tape using tweezers. A commercially available two-part epoxy that had been mixed with 325 mesh (< 44 μm) tungsten powder to provide added shielding was poured into each ring and allowed to cure at room temperature for approximately 19 hours. When the epoxy had hardened, the duct tape was removed. Each sample was manually ground using silicon carbide paper and a small amount of water to expose a longitudinal section through the fuel punching. One of the samples was initially ground with ANSI 600-grit paper (nominal abrasive size 16 μm), followed by 1200-grit (6.5 μm abrasive) paper. The other sample was initially ground with 240-grit (53 μm abrasive) paper, followed by 600-grit and 1200-grit papers. Total grinding time for each sample was 5-10 minutes.

Samples were transferred out of the glove box and decontaminated. A thin coating of sputtered Pd was applied to the surface with the exposed fuel, and the samples were transferred into the scanning electron microscope for analysis.

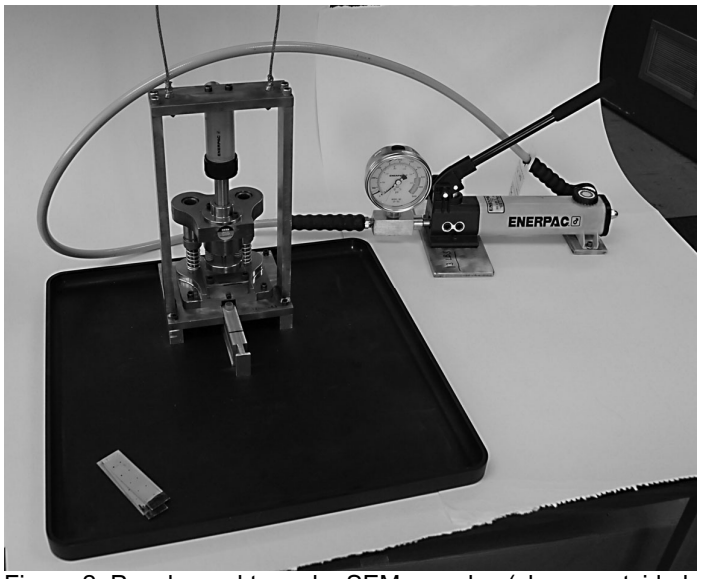

Figure 2: Punch used to make SEM samples (shown outside hot cell)

4. Results

Figure 3 shows low-magnification scanning electron microscope images of the samples. In each case, the epoxy had not completely wetted the sample, forming a bubble adjacent to the sample. As the contrast and general appearance of the sample and the tungsten-epoxy mixture are similar, these bubbles provided an easy way to locate each sample at low magnification. The tapering outline of one of the samples (Fig. 3a) suggests that the ground surface is at an angle to the long axis of the punching, while the consistent width of the other (Fig. 3b) suggests that the ground surface is approximately parallel to the long axis.

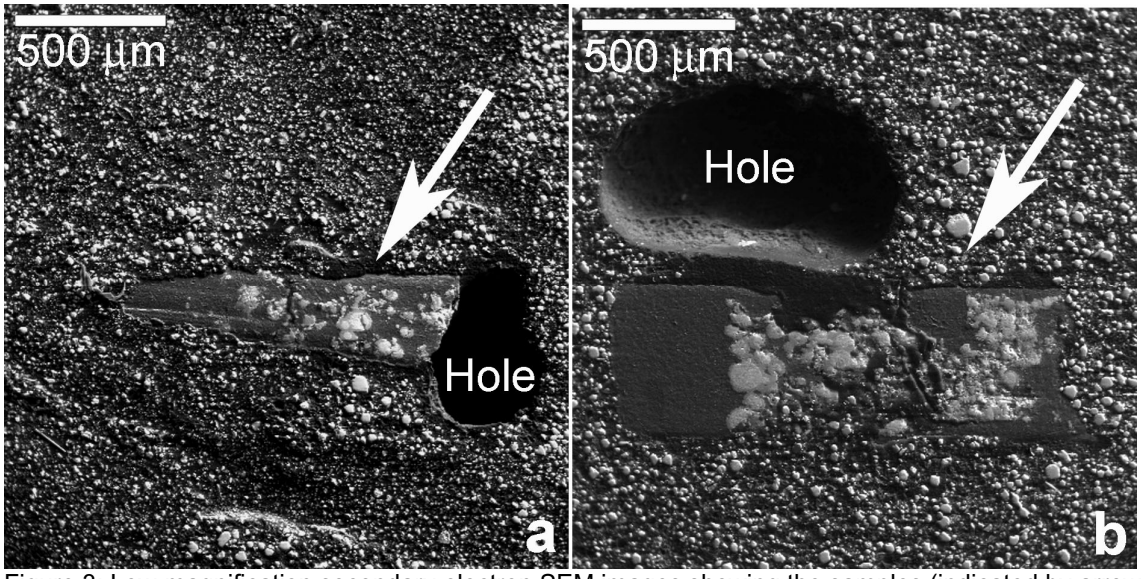

Figure 3: Low-magnification secondary-electron SEM images showing the samples (indicated by arrows) and adjacent holes. Light-colored areas in the remainder of the sample are tungsten particles embedded in the epoxy to provide shielding. a) Sample 1. b) Sample 2.

Closer inspection indicates that the samples have been strongly deformed (Fig. 4). Although the particle-free areas at the left ends of the fuel samples in Figure 3 may be cladding, the region with fuel particles extends almost to the right ends of both of the fuel samples, suggesting that the cladding at these ends of the samples

may have broken off. Sample 2 is narrower through the fuel zone than at either end (Fig. 4a), and both fuel samples show cracks extending across the punchings (Fig. 4a, b). Each sample shows regions in which sharp-edged fragments of what appear to be different materials have been jumbled together (Fig. 4c), and each shows particle-free high-Al areas along the outer edges of the punchings (Fig. 4d).

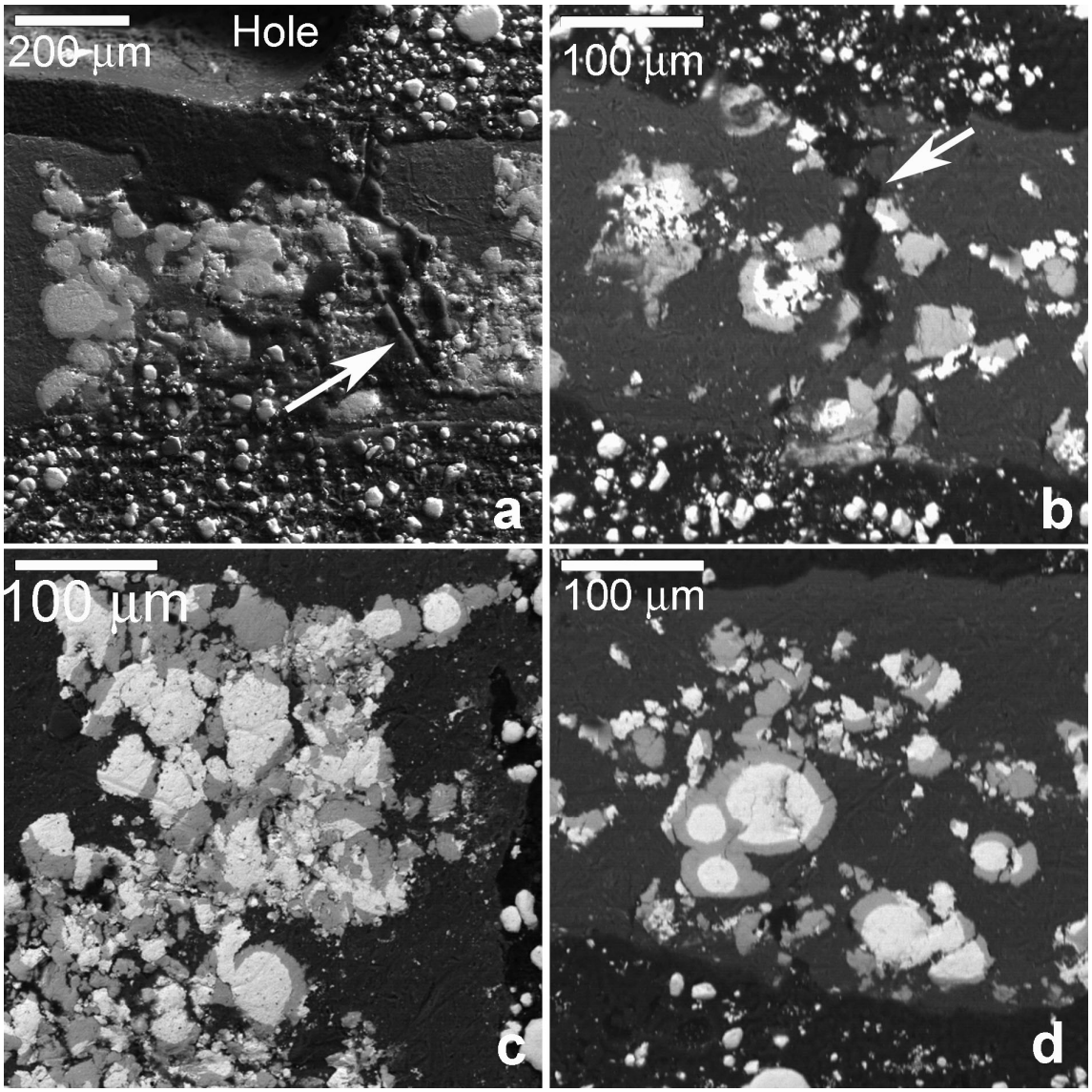

Figure 4: Evidence for deformation of fuel punchings. a) Narrowing in fuel zone and crack, Sample 2. b) Crack, Sample 1. c and d) Fuel-free zones along edges of punchings and jumbled fragments of fuel particles and reaction rims, Samples 2 and 1, respectively.

None of these characteristics would be expected from perfect cylinders taken from fuel plates with microstructures similar to those in Figure 1. However, higher-magnification images show a few particles with concentric microstructures that resemble those in the post-irradiation optical images (Fig. 5). X-ray maps confirm that the light-colored centers of these particles are high in U and Mo, the intermediate-contrast areas around the outsides are high in U and Al, and the dark colored material surrounding the particles is predominantly Al. Thus, these particles can plausibly be interpreted as representing remnants of an original fuel particle surrounded by at least the inner portion of the layer of reaction products that formed around it.

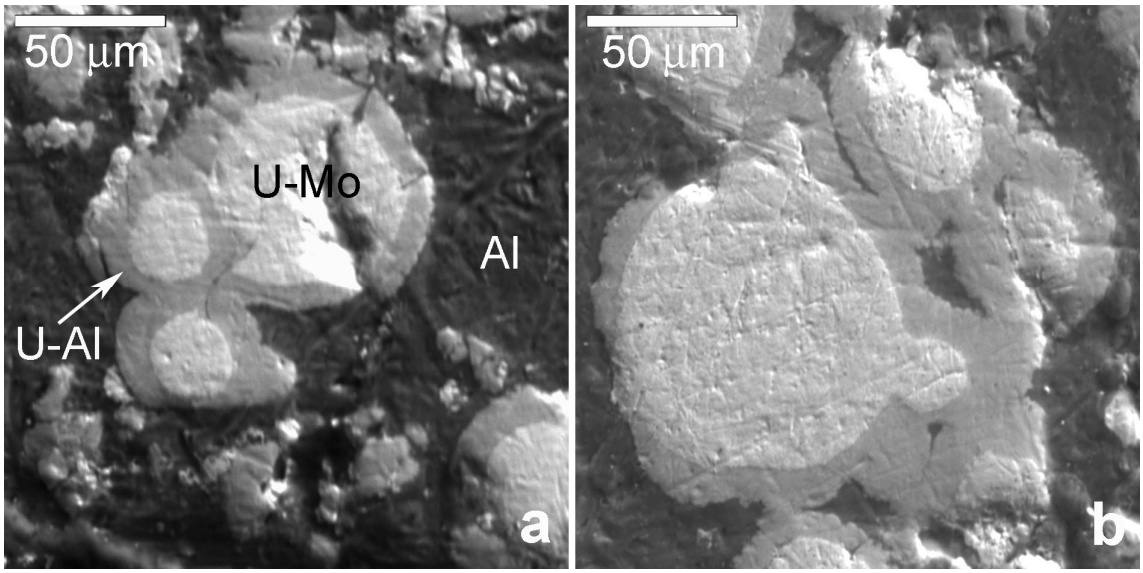

Figure 5: Secondary-electron SEM images showing microstructures similar to those in optical images. U-Mo, U-Al, and Al indicate major elements present in each area, as determined from X-ray maps. a) Sample 1. b) Sample 2.

4. Discussion

Comparisons between Figures 1, 3, 4, and 5 clearly show that the microstructure of the irradiated fuel has been changed during SEM sample preparation. Localized fractures such as those shown in Figures 3, 4a, and 4b are to be expected in brittle materials, and are unlikely to significantly affect microstructures not immediately adjacent to the fractures. However, necking of the sample in the fuel zone (Fig. 4a), particle-free areas at the edges of the punchings, and jumbled fragments (Fig. 4b-d) suggest more pervasive deformation throughout the sample, with possible flowing of the aluminum matrix. As the embedding and grinding processes used to prepare the optical and SEM samples are similar, it seems likely that the vast majority of the deformation occurred during punching.

Because of the pervasive deformation, extreme caution is necessary in interpreting data from the matrix, or data involving spatial relationships in materials in jumbled areas. Despite the pervasive deformation, it is possible to find particles whose microstructures resemble those in the optical images from the relatively intact plates. X-ray maps from these particles show chemical distributions similar to those previously reported from samples prepared with other techniques (e.g., [2-6]), and it may be appropriate to ignore the deformation of the surrounding material in interpreting data from these particles. However, data from these particles should always be compared to that obtained from samples prepared by other techniques.

Decisions about how to prepare samples and perform electron-microscope analysis of irradiated fuel necessarily involve considerations of convenience, cost, and safety. One could no doubt obtain excellent data by putting an entire cross-section of a fuel plate similar to that in Figure 1 into a scanning electron microscope. But, that would require either putting an electron microscope into a hot cell or handling relatively large volumes of fuel outside a hot cell. Both of these approaches have significant problems: putting a microscope into a hot cell leads to increased costs because of difficulty in maintaining an instrument or the need to replace it relatively frequently, while handling large volumes of fuel outside a hot cell requires great care in addressing radiation safety problems.

The approach taken here provides a compromise, in which all manipulations involving large amounts of material are performed in a hot cell, while scanning electron microscopy is perfumed outside the cell. Although the results appear promising, the data suggest that starting with slightly larger cylinders of material (e.g., a 1/16”=1.65 mm punching instead of the 1 mm used here) might produce significantly larger areas with microstructures comparable to those in optical images. Nonetheless, researchers must be constantly aware of possible deformation, particularly in the matrix where it may produce little or no direct microstructural evidence.

Comparisons between radiation readings measured on the sides of the sample with the fuel punchings and those measured in an orientation in which the tungsten-epoxy mixture was between the meter and the punchings

indicated that the tungsten-epoxy mixture reduced the beta-gamma radiation from the sample by at least two orders of magnitude.

The present data illustrates both the potential and the problems associated with samples prepared from fuel-plate punchings, and suggest that further work using larger-diameter punchings may be worthwhile. Relatively minor modifications of the techniques presented here, such as using a core drill instead of a punch, should be considered.

Acknowledgments

This work was supported by the U.S. Department of Energy, Office of Nuclear Materials Threat Reduction (NA-212), National Nuclear Security Administration, under DOE-NE Idaho Operations Contract DE-AC07-05ID14517. The authors would like to thank Dr. Dan Wachs (US Technical Lead for RERTR) and Dr. Dennis D. Keiser, Jr. for their helpful comments and Mr. Duane Atwood for his assistance with radiological work permits.

References

1. D.M. Wachs, D.D. Keiser, Jr., M.K. Meyer, D.E. Burkes, C.R. Clark, G.A. Moore, J.F. Jue, M.R. Finlay, T. Totev, G.L. Hofman, T. Wiencek, Y.S. Kim, and J.L. Snelgrove. Progress in the development of LEU fuel, in: Global 2007, Boise, ID, American Nuclear Society, 2007.
2. A. Leenaers, S. Van den Berghe, E. Koonen, C. Jarousse, F. Huet, M. Trotabas, M. Boyard, S. Guillot, L. Sannen, and M. Verwerft, Post-irradiation examination of uranium-7 wt% molybdenum atomized dispersion fuel, Journal of Nuclear Materials, 335 (2004) 39-47.
3. M.K. Meyer, G.L. Hofman, R.V. Strain, C.R. Clark, and J.R. Stuart. Metallographic analysis of irradiated RERTR-3 fuel test specimens, in: RERTR, 2000.
4. J.M. Park, H.J. Ryu, Y.S. Lee, D.B. Lee, S.J. Oh, B.O. Yoo, Y.H. Jung, D.S. Sohn, and C.K. Kim. An investigation on the irradiation behavior of atomized U-Mo/Al dispersion rod fuels, in: RERTR 2004, 2004
5. G.L. Hofman, Y.S. Kim, M.R. Finlay, J.L. Snelgrove, S.L. Hayes, M.K. Meyer, C.R. Clark, and F. Huet. Recent observations at the postirradiation examination of low-enriched U-Mo miniplates irradiated to high burnup, in: RRFM, 2004.
6. F. Huet, J. Noirot, V. Marelle, S. Dubois, P. Boulcourt, P. Sacristan, S. Naury, and P. Lemoine. Post irradiation examinations on UMo full sized plates: IRIS2 experiment, in: RRFM, 2005.